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<p>(21) International Application Number: PCT/US92/01443 (22) International Filing Date: 24 February 1992 (24.02.92) (30) Priority data: 660,402 22 February 1991 (22.02.91) US (71) Applicant: EXXON CHEMICAL PATENTS INC. [US/ US]; 1900 East Linden Avenue, Linden, NJ 07036-0710 (US). (72) Inventors: MEHTA, Aspy, Keki ; 5611 Forest Timbers, Humble, TX 77346 (US). CHEN, Michael, Chia-Chao ; Overijse Steenweg 66, B-3080 Duisburg-Tervuren (BE).</p>	<p>(74) Agents: BUTTS, Evan et al.; Exxon Chemical Company, P.O. Box 5200, Baytown, TX 77522-5200 (US). (81) Designated States: AT (European patent), BE (European patent), BR, CA, CH (European patent), DE (European patent), DK (European patent), ES (European patent), FR (European patent), GB (European patent), GR (Eu- ropean patent), IT (European patent), JP, KR, LU (Eu- ropean patent), MC (European patent), NL (European patent), SE (European patent). Published <i>Without international search report and to be republished upon receipt of that report.</i></p>	
<p>(54) Title: HEAT SEALABLE BLEND OF VERY LOW DENSITY POLYETHYLENE OR PLASTOMER WITH POLY- PROPYLENE BASED POLYMERS AND HEAT SEALABLE FILM AND ARTICLES MADE THEREOF</p>		
<p>(57) Abstract</p> <p>Heat sealable compositions suitable for film and film structures comprise: (a) from 30 to 70 weight percent of a low melt- ing polymer comprising an ethylene based copolymer having a density of from 0.88 g/cm³ to 0.915 g/cm³, a melt index of from 1.5 dg/min to 7.5 dg/min, a molecular weight distribution no greater than 3.5, and a composition distribution breath index greater than 70 percent; and, (b), being different from (a), from 70 to 30 weight percent of a propylene based polymer hav- ing from 88 mole percent to 100 mole percent propylene and from 12 mole percent to 0 mole percent of an alpha-olefin other than propylene.</p>		

HEAT SEALABLE BLEND OF VERY LOW DENSITY POLYETHYLENE
OR PLASTOMER WITH POLYPROPYLENE BASED POLYMERS AND
HEAT SEALABLE FILM AND ARTICLES MADE THEREOF

5

BACKGROUND OF THE INVENTION

1. Field of the Invention

This invention relates to blends of very low density polyethylene or plastomers with propylene based polymers. In another aspect, this invention relates to blends of very low density polyethylenes or plastomers with propylene based polymers which are useful as heat sealable compositions, and to films and articles made thereof.

2. Description of the Prior Art

Plastic films have found utility in a wide variety of packaging applications such as for example bags, pouches, tubes and trays. In many film packaging applications it is necessary to seal the film during the packaging operation. This may be accomplished by using adhesives or by using heat sealing techniques. When heat sealing is used, it is important that the plastic film be readily heat sealable while also possessing other good physical and mechanical properties such as resistance to tearing, tensile strength, and processability in high speed equipment.

For example in packaging items into a bag made from film, film is generally first processed into a bag, a bottom being formed by squeezing together two films, and subjecting the bottom to a temperature above the seal initiation temperature under pressure, to seal the bottom of the bag. The bag is subsequently filled with the goods to be packaged, and the top is sealed in a fashion similar to how the bottom was sealed.

Film heat sealing is generally affected by means of heated flat surfaces, between which the films

polypropylene copolymers) typically require heat sealing initiation temperatures upwards of about 120°C before adequate film seal strengths (at least 200 g/inch, desirably 400 g/inch and higher per specified settings for pressure and dwell time) are obtained. Consequently, there has been quite a bit of development work to find ways that would allow the heat sealing of propylene films at lower temperatures. Such approaches have included the use of coatings, blend components and multiple film layers. For example, U.S. Patent No. 4,291,092 and divisionals, U.S. Patent Nos. 4,339,496, 4,340,640 and 4,340,641, all disclose a heat sealable packaging film layer for a polypropylene substrate wherein the film layer comprises a blend of a copolymer of ethylene and a higher olefin and a copolymer of propylene and a higher olefin. U.S. Patent No. 4,643,945 discloses the use of a linear low density polyethylene in a heat sealable film composition, and E.P. 0221726 discloses a heat sealable layer comprising very low density polyethylene blended optionally with a polyolefin.

Unfortunately, the prior art heat sealable films are not without certain deficiencies. A need still exists in the industry for a heat sealable layer having a seal initiation temperature of about 110°C or lower while the film maintains good elevated temperature resistance, abrasion resistance, blocking resistance, good strength and rigidity, and good film optical properties.

A class of highly active olefin catalysts known as metallocenes is well known especially in the preparation of polyethylene and copolyethylene-alpha-olefins. These catalysts, particularly those based on group IV B transition metals, zirconium, titanium and hafnium, show extremely high activity in ethylene

physical properties of the polymer. The type and amount of comonomer affects the physical properties and crystallizability of the copolymer. All of these structural features (MW, MWD, CD, comonomer type and amount) are readily controllable through the use of metallocene catalysts as exemplified in U.S. Patent No. 4,937,299 cited above.

Metallocene catalysts are particularly attractive in making tailored ultrauniform and super random specialty copolymer. For example, if a lower density copolymer is being made with a metallocene catalyst such as very low density polyethylene, (VLDPE), an ultrauniform and super random copolymerization will occur, as contrasted to the polymer produced by copolymerization using a conventional Ziegler catalyst.

In view of the ongoing need for heat sealable films of optimum quality for particular uses, it would be desirable to provide compositions utilizing the high quality characteristics of very low density polyethylene prepared with metallocene catalysts.

Accordingly, the present invention relates to polyolefin homopolymer and copolymer blends utilizing as a low temperature polymer, ethylene based polymers, either VLDPEs or plastomers, prepared by metallocene catalysts. The blends of the present invention are particularly useful as heat sealable films in plastic packaging.

SUMMARY OF THE INVENTION

According to one embodiment of this invention there are provided polymer blend compositions comprising: (a) a low melting polymer that is an ethylene based copolymer produced with a metallocene catalyst, and (b) a propylene based

mixing equipment. Other means to achieve a well mixed blend will be apparent to those skilled in the art.

Another embodiment of this invention comprises film or sheet articles formed from a polymer blend as above described.

According to yet another embodiment of this invention there are provided multiple layer heat sealable films comprising at least one base layer and at least one heat sealable layer, the heat sealable layer comprising a film or sheet formed from a polymer blend as described above.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 shows idealized DSC melting behavior curves for determining the proper components for a blend, wherein curve (A) represents a propylene based polymer useful as blend component (b); curve (B) represents a low melting ethylene based copolymer useful as blend component (a); curve (C) represents a polymer blend of components (a) and (b); and curve (D) represents a low melting ethylene based copolymer which does not have an "essentially single" melting point in the range of 60 to 125°C and is not suitable as a blend component (a).

Fig. 2 is a graph of melting point vs. density for various copolymers of ethylene; circle, diamond and triangle points representing copolymer materials produced by a non-metallocene catalyst, and box points and line representing copolymer materials produced by a metallocene catalyst.

Fig. 3 is a DSC curve for a low melting ethylene based copolymer which does not have an essentially single melting point in the range of 60 to 125°C and is not suitable as a blend component (a).

component (a); the component (a) being the polymer of Fig. 4.

DETAILED DESCRIPTION OF THE INVENTION

5 The polymer blend compositions of the present invention that are useful as a heat sealable composition for making heat sealable polypropylene films and other heat sealable articles comprise (a) a low melting polymer ingredient that is an ethylene
10 based copolymer produced with a metallocene catalyst; and (b) a propylene based homopolymer or copolymer, which is different from (a).

 The term "PLASTOMERS" as used herein refers generally to a class of ethylene based copolymers with
15 density of less than about 0.900 g/cm^3 (down to about 0.865 g/cm^3) at a molecular weight, M_w greater than about 20,000 (about 200 MI and lower). Plastomers have an ethylene crystallinity between PLASTics (i.e. linear low density and very low density polyethylenes)
20 and ethylene/alpha-olefin elastOMERS.

 In contrast, VLDPEs typically have densities in the range of about 0.90 to about 0.915 g/cm^3 .

 Other properties of the plastomers and VLDPEs suitable for use in the present invention are
25 explained herein.

 The preference as to whether to use VLDPE's or plastomers as the low melting polymer ingredient will depend on the requirements of the particular application in which the blend will be utilized,
30 economics, and other process factors.

 The compositions of the present invention comprise from about 30 to about 70 percent, preferably 40 to about 60, and most preferably about 50 weight percent low melting polymer, and from about 70 to
35 about 30 percent, preferably 60 to about 40, and most

The low melting polymer ingredient of the present invention should have an essentially single melting point characteristic with a peak melting point (T_m) as determined by Differential Scanning Colorimetry (DSC) in the range of about 60°C to about 115°C. Preferably the DSC peak T_m is in the range of about 80°C to about 100°C. "Essentially single melting point" as used herein means that at least about 80% wt of the material corresponds to a single T_m peak existing in the range of about 60-115°C, and there is essentially absent from the polymer any substantial fraction of material which corresponds to a T_m peak found at a temperature higher than about 115°C, i.e., "essentially" the bulk material content of the polymer corresponds to a "single" melting point peak in the 60-115°C range, and "essentially" no substantial fraction of the material has a peak melting point in excess of about 115°C, as determined by DSC analysis.

DSC measurements were made on a Perkin Elmer System 7 Thermal Analysis System. Melting information reported are second melting data, i.e., sample heated at programmed rate of 10°C/min to a temperature above its melting range. Sample then cooled in instrument at programmed rate of 10°C/min to temperature below its crystallization range. Sample reheated (2nd melting) at programmed rate of 10°C/min.

The presence of higher melting peaks is detrimental to film properties such as haze, and compromises the chances for meaningful reduction in the seal initiation temperature of the final film.

Fig. 1A shows the Melting behavior for neat random copolymer polypropylene. The peak melting point shown is approximately 132°C which translates to

would deteriorate the optical properties of the random copolymer blend film (e.g., cause increase in film haze). The melting of the blend would be a superposition and merging of the melting behaviors shown for the individual components.

Fig. 1D shows the melting behavior of standard Zeigler-catalyzed VLDPEs as the low melting component (schematic for typical 0.908 D VLDPE). These products are generally broader in molecular weight distribution and composition distribution and are less desirable as the low melting blend component. They are almost always characterized by multiple melting peaks (2 or more), particularly peaks of higher melting temperature, reflective of the high ethylene containing (high crystallinity) species in the composition. The presence of these high melting peaks is not significantly influenced by lowering product density.

Data measurements over many samples, depicting this feature, are shown in Figure 2 which shows the high crystallinity species that cause undesirable effects. Their high melting behavior (peak positions are around 120 to 125°C, which is not far removed from that of the random copolymer polypropylene) significantly compromised the chances for a meaningful reduction in the polypropylene's seal initiation temperature, and they contribute to an increase in film haze. While some reduction in the relative amount of these high melting species is possible by going to lower overall product densities, this approach increases the content of amorphous, extractable species, which is undesirable in food contact applications.

The percent hexane extractables for the low melting polymer ingredient of the present invention

above 70%) from VLDPEs available commercially today (broad composition distribution as assessed by CDBI values generally less than 55%). The benefits to the subject invention that accrue through the specific use of VLDPEs/plastomers of narrow composition distribution are elucidated later in the examples. The CDBI of a copolymer is readily calculated from data obtained from techniques known in the art, such as, for example, temperature rising elution fractionation as described, for example, in U.S. Patent Application Serial No. 131,350, U. S. Application Serial No. 151,350, filed February 2, 1988, or in Wild et al., J. Poly. Sci, Poly. Phys. Ed., vol. 20, p. 441 (1982). Unless otherwise indicated, terms such as "comonomer content", "average comonomer content" and the like refer to the bulk comonomer content of the indicated interpolymer blend, blend component or fraction on a molar basis.

Utilizing a metallocene catalyst, the VLDPEs and plastomers useful as the low melting polymers of the present invention can be produced in accordance with any suitable polymerization process, including a slurry polymerization, gas phase polymerization, and high pressure polymerization process.

A slurry polymerization process generally uses super-atmospheric pressures and temperatures in the range of 40-100°C. In a slurry polymerization, a suspension of solid, particulate polymer is formed in a liquid polymerization medium to which ethylene and comonomers and often hydrogen along with catalyst are added. The liquid employed in the polymerization medium can be an alkane, cycloalkane, or an aromatic hydrocarbon such as toluene, ethylbenzene or xylene. The medium employed should be liquid under the

temperature of said product and that the polymerization pressure be above about 500 bar (kg/cm²). In those situations wherein the molecular weight of the polymer product that would be produced at a given set of operating conditions is higher than
5 desired, any of the techniques known in the art for control of molecular weight, such as the use of hydrogen or reactor temperature, may be used in the process of this invention.

10 Polypropylene random copolymer is preferred over homopolypropylene since it has a lower melting temperature and lower seal initiation temperature. The concept of blending a low melting ingredient like VLDPE or plastomer would however also apply to
15 homopolypropylene.

The propylene based polymers or copolymers of the present invention are generally any propylene based polymer or copolymer of propylene such as
20 homopolypropylene, impact copolymers, polypropylene copolymers, and polypropylene random copolymers. Preferably the propylene base polymer or copolymer is homopolypropylene or polypropylene random copolymers. Most preferably, the propylene based polymer or
25 copolymer is polypropylene random copolymer.

The polypropylene random copolymer useful in the heat sealable blend composition of the present invention is different from the polyethylene based
30 copolymer component and is composed of propylene as a main component and an alpha-olefin other than propylene. The content of the propylene in the random copolymer is generally in the range of about 88 to about 99 mole percent, based on total moles, preferably in the range of about 90 mole percent to about 94 mol percent. The content of the alpha-olefin
35 (e.g. ethylene) other than propylene is generally in

propylene based polymer, it is believed that a deterioration of haze can be mitigated by blending low melting polymer ingredient and propylene based polymer having similar densities to minimize refractive index differences, and having similar rheologies (i.e., shear viscosities under processing conditions) to obtain optimum dispersed phase domain size/morphology.

The heat sealable blend composition of the present invention can be used to manufacture heat sealable film and articles using film fabrication technologies well known to those skilled in the art. For example, the blend may be cast extruded or coextruded into film using a flat die or blown extruded or coextruded into film using a tubular die. Such films may optionally be further oriented (either uniaxially or biaxially) using technologies well known to those skilled in the art. The heat sealable film and articles of the present invention may be incorporated onto film substrates or other articles to provide for heat sealing. For example a heat sealing film of the present invention may be included onto one or both surfaces of a prefabricated base layer of substrate film via lamination. The heat sealable film(s) or base substrate film may themselves be un-oriented, uniaxially oriented or biaxially oriented.

The base substrate layer or film will most commonly be polypropylene (homo or co-polymer). Other polyolefin type substrates however could also benefit from the heat sealable blend compositions of this invention.

The above are just a few examples of film constructions utilizing the heat sealable blend composition of this invention. Several others are possible, as will be apparent to those skilled in the art of film construction design and fabrication.

Table I

Gas Phase Polymerization

	Temperature (°F)	144
	Total Pressure (psia)	300
5	Gas Velocity (ft/sec)	1.59
	Catalyst Feed Rate (g/hr)	4.8
	Production Rate (lb/hr)	40

The polymerized product had a weight average molecular weight (M_w) of 87,000 and a polydispersity (M_w/M_n) of 2.05. The density was measured as 0.897 g/cm³.

EXAMPLE II

Narrow molecular weight distribution and narrow composition distribution VLDPEs/Plastomers as the low melting ingredient in the blend, versus currently available VLDPEs, show several advantages. The narrower CD provides a lower melting temperature, for lower seal initiation temperature and better tailoring of the melting point distribution of the blend product, to provide better low temperature sealing capability. Additionally, the lower extractables permit broader food contact possibilities, and better blocking resistance. The narrower MWD enhances the haze/clarity properties of a film. A comparison of these resin/film properties for neat VLDPE of the type required and used in this invention versus competitive, more broadly distributed VLDPEs is shown in Table II.

The competitive VLDPEs listed in Table II were selected on the basis of their density being the lowest density material which is commercially available. From Mitsubishi an ethylene-1-butene was available, whereas the lowest density material available from Dow was an ethylene-1-octene material. The DSC curve for the Dow material is shown in Fig. 3.

Table II
 RESIN/FILM PROPERTIES COMPARISON*
 NEAT VLDPE USED IN THIS INVENTION V.
COMPETITIVE, MORE BROADLY DISTRIBUTED VLDPEs

5	<u>SAMPLE</u>	<u>COMPETITIVE</u> <u>VLDPE A</u>	<u>COMPETITIVE</u> <u>VLDPE B</u>	<u>VLDPE-1</u> <u>VLDPE AS</u> <u>DESCRIBED IN</u> <u>THIS INVENTION</u>
10		[Mitsubishi x-142]	[Dow ATTANE 4003]	
	<u>COMONOMER</u>	C ₄	C ₈	C ₄
15	MI [dg/min]	0.9	0.8	1.7
	Density [g/cm ₃]	0.900	0.906	0.897
20	M _w /M _n	3.1	3.7	2.0
	DSC Peak T _m s [°C]	91,110	96,123	77,102
25	Composition Distribution Breadth Index [CDBI%]	55	46	75
30	[NOTE: A higher value of breadth index implies a narrow Composition Distribution]			
35	Hexane Extract [%]	8	4.9	3.5
40	Haze [%] (1.25 mil blown film)	5	10.5	3.4
45	<u>CAPSULE:</u>	THE NARROWER MOLECULAR WEIGHT DISTRIBUTION AND COMPOSITION DISTRIBUTION OF THE VLDPEs AND PLASTOMERS OF THIS INVENTION VS THE COMPETITIVE VLDPEs ALLOWS;		
		- LOWER DSC PEAK MELTING TEMPERATURE(S)		
		- LOWER FILM HAZE		
		- LOWER HEXANE EXTRACTABLES		

copolymer polypropylene) film is shown in Figure 9 for comparison.

The addition of VLDPE-1 to random copolymer polypropylene allows sealing to occur at lower
5 temperatures than is possible with neat random copolymer polypropylene by itself. The blends provide a good balance of sealability at lower temperatures coupled with high seal strength.

EXAMPLE IV

10 Preparation of VLDPE-2 (2.1 MI 0.906, Butene-1-Comonomer)

The catalyst for polymerizing this ethylene copolymer was prepared as follows. An 800 gram quantity of silica gel and a 2700 ml aliquot of
15 methylalumoxane/toluene solution (10%) were placed in a two-gallon reactor and allowed to react at ambient temperature for one hour. A 21.6 gram quantity of di-(n-butylcyclopentadienyl) zirconium dichloride slurried in 300 ml of toluene was added into the
20 reactor and the mixture was allowed to react at 65°C for 30 minutes. The reactor was then heated at 75°C while nitrogen gas was purged through the reactor to remove the solvent. The heating and nitrogen purging were stopped with the mixture in the reactor turned
25 into a free-flowing powder.

The polymerization was conducted in a 16-inch diameter fluidized bed gas-phase reactor. Ethylene, butene-1, and nitrogen were fed continuously into the reactor to maintain a constant production
30 rate. Product was periodically removed from the reactor to maintain the desired bed weight. The polymerization conditions are shown in the Table below.

A catalyst is prepared by adding 5.1 liters of a 10% solution of trimethylaluminum in heptane into a dry and oxygen-free two-gallon reactor equipped with a mechanical stirrer. 800 g of undehydrated silica gel, containing 12.3% water, is slowly added into the reactor. After the addition is complete, the mixture is stirred at ambient temperature for one hour. 20 g of di-(n-butylcyclopentadienyl) zirconium dichloride slurried in 30 ml of heptane is then added into the reactor and the mixture is allowed to react at ambient temperature for 30 minutes. The reactor is then heated to 65°C, while a nitrogen gas is purged through the reactor to remove the solvent. The nitrogen purging is stopped when the mixture in the reactor turns into a free-flowing powder.

The polymerization was conducted in a 16-inch diameter fluidized gas phase reactor. Ethylene, butene-1 and nitrogen were fed continuously into the reactor to maintain a constant production rate. Product was periodically removed from the reactor to maintain the desired bed weight. The polymerization conditions are shown in the table below.

the blends and neat PD-9282 random copolymer polypropylene (RCP) are shown in Figure 12.

The data shows:

- 5 - RCP/VLDPE blends show good extractables levels, lower than neat RCP. Blends with the Plastomer-1 however show higher extractables.
- 10 - DSC melting curves show the capability to extend the melting range of RCP to lower temperatures, via the addition of VLDPE or Plastomer. Particularly for blends of RCP with VLDPE [density in the range of about 0.90-0.91 g/cm³], good merging of the individual melting ranges was noted, which is believed to be desirable [see Fig. 12].
- 15 - Haze/Gloss numbers on the blend films are, to a first approximation, in line with the neat RCP film values.

20 In review, the data show that a narrow molecular weight distribution and narrow composition distribution VLDPE (density of about 0.90-0.91 g/cm³) provides an improved balance of properties in blends with random copolymer polypropylene: strong, lower temperature seals; low extractables; low film haze.

EXAMPLE IX

Coextruded oriented polypropylene (OPP) films were produced using blends of VLDPE and random copolymer polypropylene as skin layers. The films were produced on a pilot OPP film line, producing biaxially oriented films. The films were 3 layer coextruded films, with skin layers on either side of a homopolypropylene core (1 μ /20 μ /1 μ thicknesses). The core layers were Exxon's Escorene™ PP-4252 (3 MFR propylene homopolymer). The preparation of coextruded, biaxially oriented films is readily done using techniques well known in the art. Polymer is extruded through a slot die in sheet form, which is then oriented sequentially along the machine and transverse directions at elevated temperatures. Typical values set for some key processing parameters were:

	Melt temperature	265°C
	MD Stretching Temperature	100°C
20	MD Stretching Ratio	5.5
	TD Stretching Temperature	160°C
	TD Stretching Ratio	8.0

Property data on two films is shown in Table VI, which had constructions as follows:

25	<u>Film 1</u>	
	Core layer:	PP-4252 (3 MFR homopolymer)
	Skin layer:	blend of 80% PD-9282 (random copolymer polypropylene; 5 MFR, 5 wt% ethylene comonomer) with 20% VLDPE 1 (1.7 MI, 0.897D, butene-1 comonomer)
30		

Table VI

VLDPE/RANDOM COPOLYMER POLYPROPYLENE BLEND OPP FILMS

5	FILM	SKIN LAYER		SEAL STRENGTH, g/in				TOTAL	
		(COMPOSITION %)						HAZE	GLOSS
		RCP	VLDPE	110°C	120°C	130°C	140°C	%	%
10	FILM 1 (Invention)	80	20*	100	341	363	381	4	72
	FILM 2 (Comparative)	80	20**	10	236	286	318	19	40
15	*	VLDPE-1 [1.7 MI, 0.897D, butene-1 comonomer; narrow MWD and CD]							
20	**	VLDPE-B [0.8 MI, 0.906D, octene-1 comonomer; broader MWD and CD]							

comprising a cyclopentadienyl-transition metal compound, preferably a metallocene such as a titanocene, zirconocene, or hafnocene, optionally in combination with an alumoxane cocatalyst.

5 9. The composition of claim 1 wherein (b) is a polypropylene random copolymer.

10 10. The composition of claim 9 wherein (b) comprises from 90 to 94 mole percent propylene and from 10 to 6 mole percent of an olefin other than propylene.

11. A film or sheet comprising a composition according to any preceding claim.

15 12. A multiple layer heat sealable film structure comprising at least one base layer and at least one heat sealable layer, the heat sealable layer comprising a composition according to any one of claims 1 to 10 or a film or sheet according to claim 11.

20 13. The film structure of claim 12 wherein the base layer comprises a polyolefin, polyester, or polyamide.

14. The film structure of claim 13 wherein the base layer is a polypropylene.

25 15. The film structure of claim 12, 13, or 14 wherein the structure is cast extruded or coextruded using flat or tubular die.

16. The film, sheet, or structure of any of claims 11 to 15 having a seal initiation temperature of 110°C or less.

30 17. A package containing food comprising the film or sheet of claim 7 or the film structure of any of claims 12 to 16, optionally formed by a process of horizontal or vertical form-fill-and-seal.

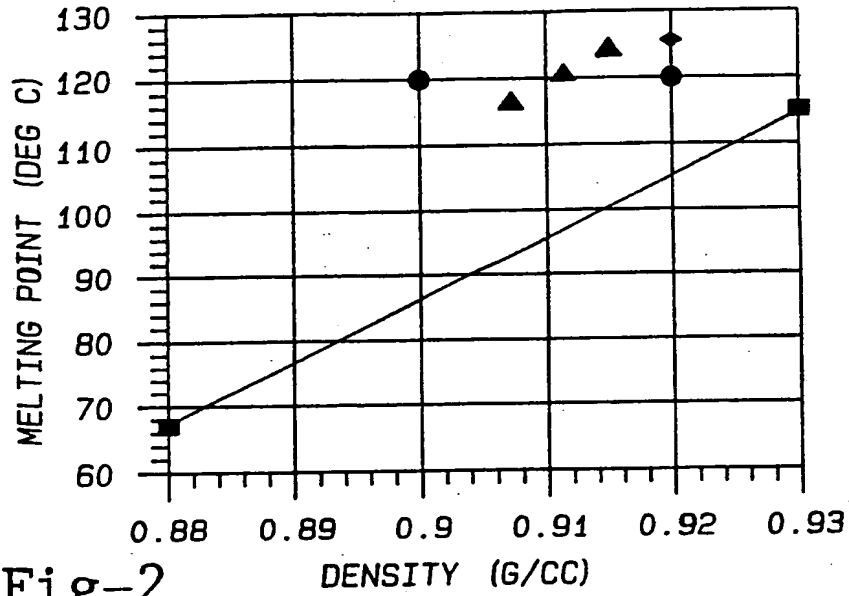


Fig-2

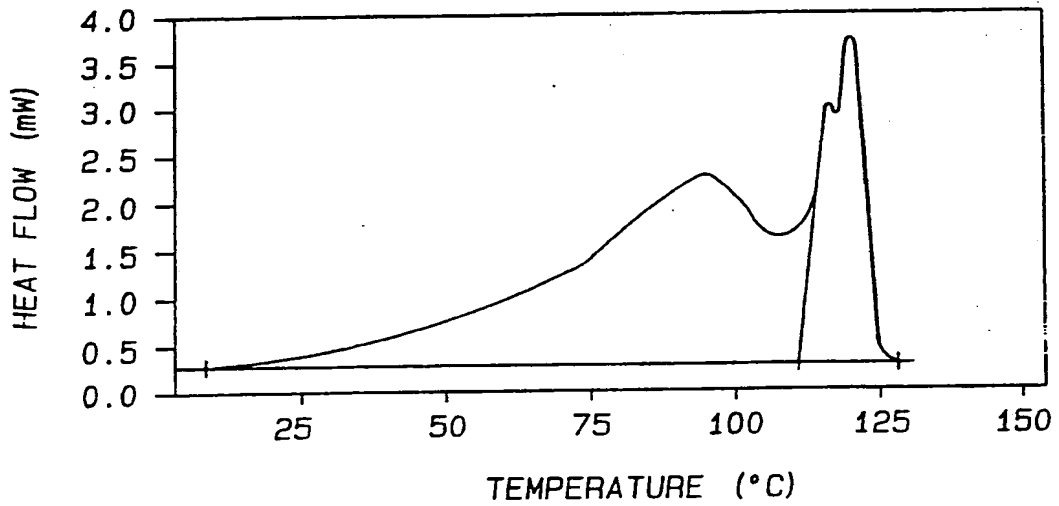


Fig-3

SUBSTITUTE SHEET

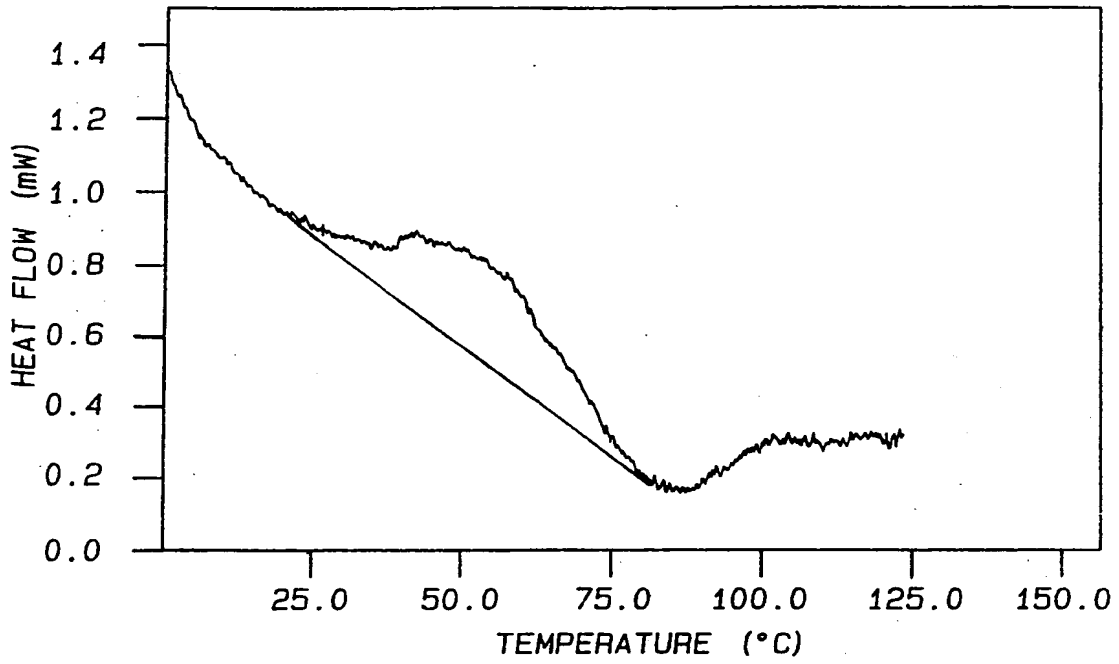


Fig-6

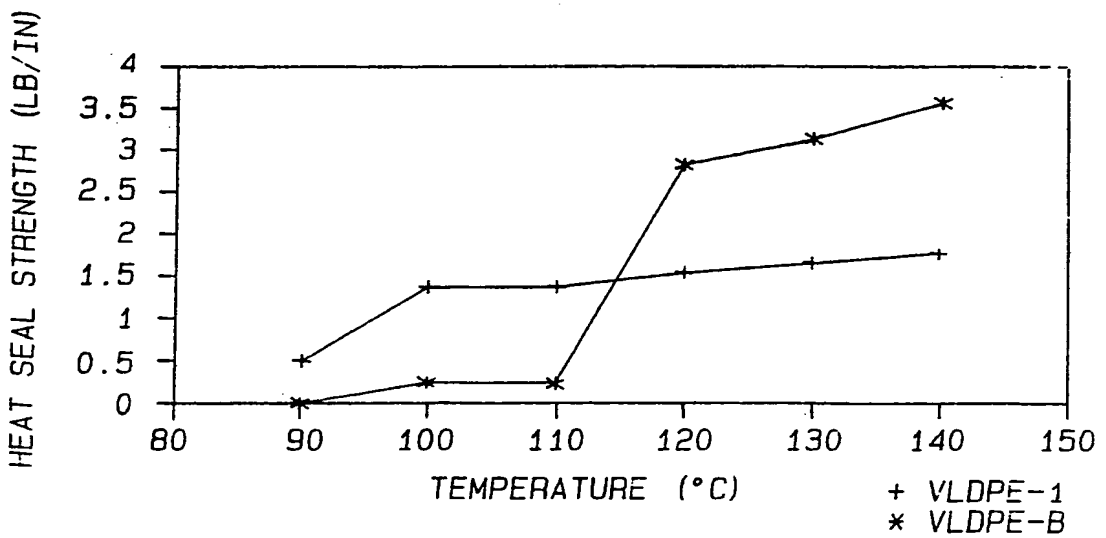


Fig-7

SUBSTITUTE SHEET

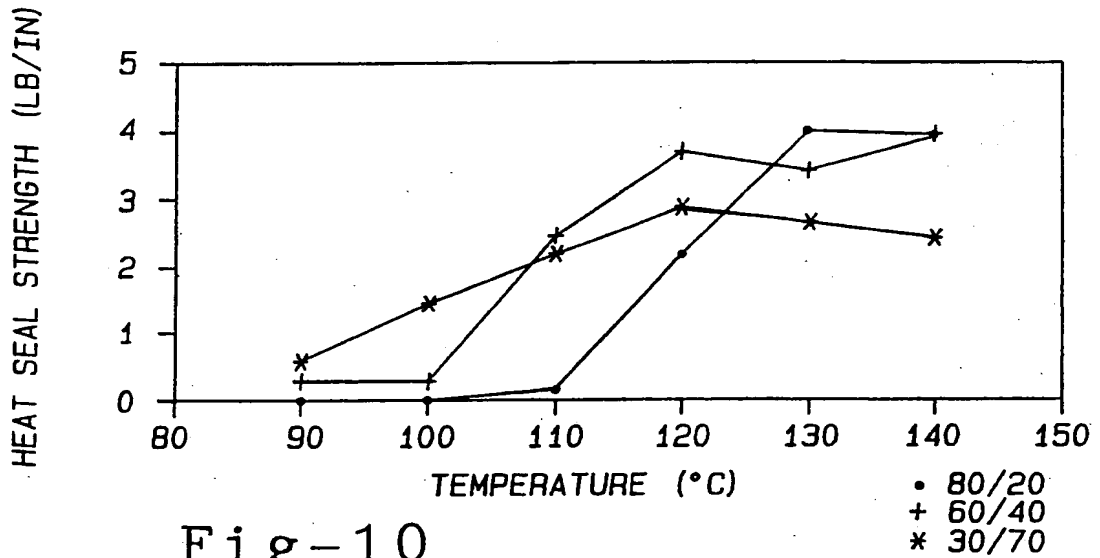


Fig-10

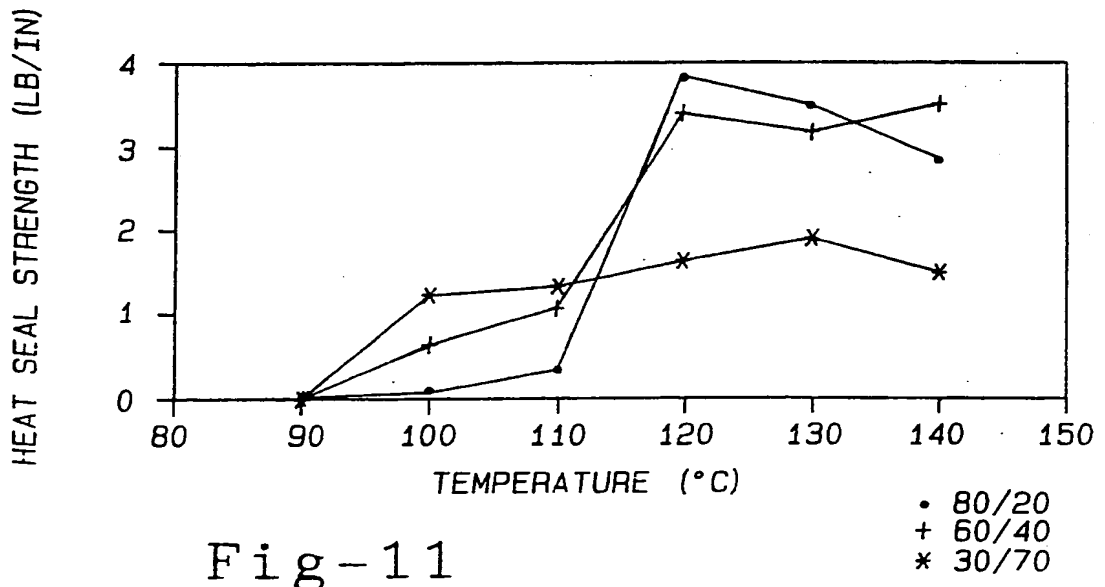


Fig-11

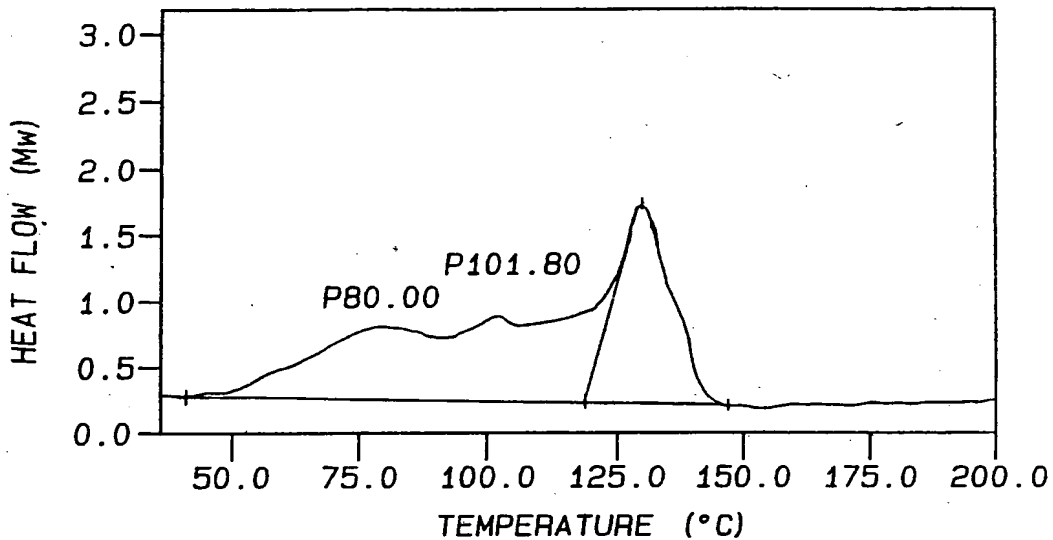


Fig-12C

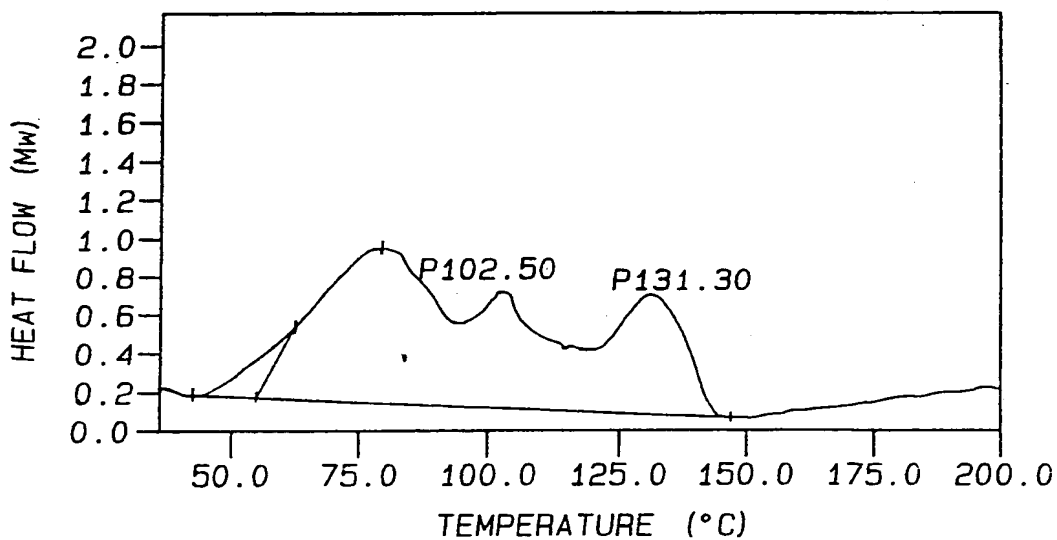


Fig-12D

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